



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of :  
Takahiro FURUTANI et al. :  
Serial No. 09/924,679 : Group Art Unit: 1714  
Filed: August 9, 2001 : Examiner: C. E. Shosho  
For: AQUEOUS INK COMPOSITION :

DECLARATION

Commissioner for Patents  
Alexandria UA 22313-1450

Sir:

I, Takahiro FURUTANI, declare that I am one of the co-inventors of the above-identified application and familiar with the prosecution history of the present application.

I declare that I have read and understand the official action dated April 12, 2004, issued against the above-identified application and also the references cited in the official action.

I declare that I have carried out the following experiments in order to demonstrate that unexpected superiority of the present invention over the cited references.

## 1. Measurement of drying time

In the present application, drying time of print is measured by use of an ink jet printer 170i manufactured by Marconi Data System Japan Co., Ltd. as described in the specification, page 24, line 17 to page 25, line 2.

However, in case of an aqueous ink composition C prepared below, clear printing was not able to be carried out so that drying time could not be measured properly. Thus, drying time was measured as follows.

By use of a wire bar No. 4 (manufactured by Toyo Seiki K.K.), each of aqueous ink compositions prepared below was printed on a PET film at a temperature of 27°C and relative humidity of 65%. After lapse of a given time, 10 g of a sash weight was placed on and rubbed the printed PET film, and the time before the print disappeared by the rubbing was measured as drying time. The shorter the drying time, the better the drying property.

## 2. Preparation of ink composition

In the following experiments, all "part(s)" means "part(s) by weight".

(1) Example 1 of the present application was followed up as described below.

Thenoyltrifluoroacetone	10.6 parts
Ethanol	274.0 parts
Europium (III) nitrate · hexahydrate	3.0 parts
Deionized water	274.0 parts

Sodium hydroxide was added to the above components under being stirred by a magnetic stirrer to adjust the pH to 6-7. Then, thereto was added 22.3 parts of polyvinyl pyrrolidone K30 (manufactured by Wako Jun-Yaku Kogyo Co., Ltd.) as a water-soluble resin, followed by stirring at 60°C for 3 hours and then filtration to obtain a phosphor solution A.

Furthermore, to the resulting phosphor complex solution A (phosphor dye from which the solvents are removed is referred to as "phosphor dye A") were added 0.2 part of BYK-348 (manufactured by BYK Chemie Co., Ltd.) as a surface treating agent and 17.0 parts of 1,2,3-benzotriazole as a quick-drying property imparting agent, followed by stirring at 25°C for 1 hour and then filtration to obtain an aqueous ink composition A.

The drying time of the ink composition A was 13 seconds.

(2) Example 3 of JP 53140105 (JP 105) was followed up as described below.

An aqueous solution of 100 mmol of europium

chloride hexahydrate in 1000 ml of water was mixed with a solution of 300 mmol of thenoyltrifluoroacetone in 150 ml of ether. After the pH of the mixture was adjusted to 8-9 with ammonia water, stirring was effected at room temperature for about 30 minutes. Then, from the mixture, water layer was isolated and removed, and the remainder (ether layer) was sufficiently washed with water. Thereafter, the remainder was dried with anhydrous sodium carbonate and filtrated, and ether in the filtrate was distilled off to prepare Eu-TTA fluorescent material.

A composition having a pH of 10.0 was prepared from 3 parts of KG-613 (styrene/maleic acid resin, trade name of Arakawa Rinsan Kagaku Kogyo K.K.), 2.9 parts of ethyleneglycol monomethyl ether, 20 parts of ethanol, 0.1 part of the Eu-TTA fluorescent material prepared above, 71 parts of distilled water and N,N-dimethylaminoethanol for adjusting pH. This composition was filtered and purified by use of a membrane filter having a pore size of  $1.0 \mu$  to obtain an aqueous ink composition B.

The drying time of the ink composition B was about 20 seconds.

(3) Example A3 of U.S. Patent No. 6,498,222 (Kitamura) was followed up as described below.

i) Preparation of Waterfastness-imparting Agent A3

A 500-ml flask equipped with a thermometer, a

stirring device, a nitrogen introduction tube, and a reflux condenser was charged with 52.6 g of N,N-dimethylaminopropylacrylamide, 23.2 g of a 75 wt % aqueous solution of (3-acrylamidopropyl)trimethylammonium chloride, 4.0 g of 2,4-diphenyl-4-methyl-1-pentene, and 130 g of N,N-dimethylformamide. A solution of 5.0 g of dimethyl 2,2'-azobis(2-methyl propionate) dissolved in 20 g of N,N-dimethylformamide was then added to the flask. The reaction mixture was heated to 80°C while stirring under a nitrogen atmosphere, and the solution was kept at 80°C for 4 hr. The reaction solution was then heated to 95°C, and the reaction was continued for additional 2 hr at 95°C, followed by cooling to room temperature to give about 230 g of a pale yellow transparent solution. The solvent was removed under the reduced pressure. A highly viscous liquid as the residue was dissolved in methyl ethyl ketone, and a precipitation procedure from a mixture of methyl ethyl ketone and hexane was repeated several times. The resulting precipitate was dried to give 40.3 g of a solid.

A 100-ml flask equipped with a thermometer, a stirring device, a nitrogen introduction tube, and a reflux condenser was charged with 20.0 g of water and 10.0 g of the solid prepared above. The reaction mixture was heated to 90°C while stirring under a nitrogen atmosphere, and the solution was then kept at 90°C for 20 hr, followed

by cooling to room temperature. Thus, about 30 g of a pale yellow transparent solution was prepared. The solvent was removed under the reduced pressure. The residue was redissolved in methyl ethyl ketone, and a precipitation procedure from a mixture of methyl ethyl ketone and hexane was repeated several times. The resulting precipitate was dried to give 9.2 g of a solid. The weight-average molecular weight and the number-average molecular weight of the polymer were determined by GPC to be 4,800 and 3,200, respectively, against polyethylene glycol standards.

ii) Preparation of ink composition C-1

To 2.0 g of the waterfastness-imparting agent A3 prepared above were added 4.5 g of C.I. Direct Black 32, 9 g of glycerin, 5 g of thiodiglycol, 5 g of diethylene glycol monobutyl ether, 10 g of N-methylimidazole, 0.9 g of triethanolamine, 0.2 g of sodium hydroxide, and 1 g of Surfynol 465 (tradename; manufactured by Air Products and Chemicals Inc.). Further, ultrapure water was added to bring the total amount of the mixture to 100 g. Thus, an aqueous ink composition C-1 was prepared.

The drying time of the ink composition C-1 was at least 30 seconds.

iii) Preparation of ink composition C-2

The same procedure as in ii) preparation of ink composition above was repeated except that 10 g of 1,2,3-

benzotriazole was used in place of 10 g of N-methylimidazole to prepare an aqueous ink composition C-2.

The drying time of the ink composition C-2 was at least 30 seconds.

### 3. Conclusion

As described above, the drying time of each of the ink compositions prepared according to the cited references is considerably longer than that of the ink composition of the present invention. As can be seen from the ink composition C-2 above, even when 1,2,3-benzotriazole, which is the quick-drying property imparting agent typically used in the present invention, is used in Kitamura, good drying property cannot be attained.

Therefore, from the teachings of the cited references, there cannot be attained the object of the present invention, i.e., to provide an aqueous ink composition which shows excellent drying property for impermeable printing materials such as plastic films without using apparatuses such as ultraviolet irradiation apparatuses and heating apparatuses and which is mild for earth environment.

Accordingly, the present invention is unobvious from the cited references even in combination.

The undersigned declarant declares further that all statements made herein of own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this                      day of Jul. 13, 2004.

Takahiro Furutani

Takahiro FURUTANI